

Benchmarking thermochemical experiments and calculations of nitrogen-containing substituted adamantanes

Emel'yanenko V., Nagrimanov R., Verevkin S.
Kazan Federal University, 420008, Kremlevskaya 18, Kazan, Russia

Abstract

© 2016, Akadémiai Kiadó, Budapest, Hungary. Standard molar enthalpy of formation of 2-cyano-adamantane was obtained by using high-precision combustion calorimetry. The standard molar enthalpies of sublimation of 2-cyano-adamantane and 2,2-dinitro-adamantane at 298.15 K were derived from the vapor pressure temperature dependences measured by transpiration. The molar enthalpies of fusion of these compounds were measured with the help of differential scanning calorimetry. Thermochemical data on functional substituted adamantanes containing nitrogen in the substituents were collected and evaluated. The gas-phase enthalpies of formation were calculated with the high-level quantum-chemical method G4 and compared with the experimental results. The consistent data set of the benchmark quality was suggested for practical thermochemical calculations. Geminal destabilizing effects for the interactions of nitro- and cyano-substituents placed in the second position on the adamantane cage were derived. Structure-property correlations for substituted adamantanes and aliphatic substituted alkanes were found and suggested for the assessment of the gas-phase enthalpies of formation of adamantane derivatives.

<http://dx.doi.org/10.1007/s10973-016-6071-9>

Keywords

Adamantane derivatives, Combustion calorimetry, Enthalpies of phase transitions, Enthalpy of formation, Quantum-chemical calculations, Transpiration method, Vapor pressure

References

- [1] De Clercq E. Antiviral drugs in current clinical use. *J Clin Virol.* 2004;30(2):115-33.
- [2] Morozov IS, Ivanova IA, Lukicheva TA. Actoprotector and adaptogen properties of adamantane derivatives (a review). *Pharm Chem J.* 2001;35(5):235-8.
- [3] Bazyleva AB, Blokhin AV, Kabo AG, Kabo GJ, Emel'yanenko VN, Verevkin SP. Thermodynamic properties of 1-aminoadamantane. *J Chem Thermodyn.* 2008;40(3):509-22.
- [4] Bazyleva AB, Blokhin AV, Kabo GJ, Charapennikau MB, Emel'yanenko VN, Verevkin SP, Diky V. Thermodynamic properties of adamantane revisited. *J Phys Chem B.* 2011;115(33):10064-72.
- [5] Fritzsche K, Dogan B, Beckhaus HD, Ruechardt C. Geminal substituents effect. Part I. Thermochemistry of 1-nitro-, 2-nitro-, 2,2-dinitro- and 2-cyano-2-nitroadamantane. *Thermochim Acta.* 1990;160:147-59.

- [6] Emel'yanenko VN, Verevkin SP, Heintz A. The gaseous enthalpy of formation of the ionic liquid 1-butyl-3-methylimidazolium dicyanamide from combustion calorimetry, vapor pressure measurements, and ab initio calculations. *J Am Chem Soc.* 2007;129(13):3930-7.
- [7] Hubbard WN, Scott DW, Waddington G. In experimental thermochemistry. In: Rossini FD, editor. Interscience Publishers. New York; 1956. pp. 75-127.
- [8] Verevkin SP, Emel'yanenko VN. Transpiration method: vapor pressures and enthalpies of vaporization of some low-boiling esters. *Fluid Phase Equilib.* 2008;266(1-2):64-75.
- [9] Verevkin SP, Sazonova AY, Emel'yanenko VN, Zaitsau DH, Varfolomeev MA, Solomonov BN, Zherikova KV. Thermochemistry of halogen-substituted methylbenzenes. *J Chem Eng Data.* 2015;60(1):89-103.
- [10] Frisch MJ, Trucks GW, Schlegel HB, Scuseria GE, Robb MA, Cheeseman JR, Scalmani G, Barone V, Mennucci B, Petersson GA, Nakatsuji H, Caricato M, Li X, Hratchian HP, Izmaylov AF, Bloino J, Zheng G, Sonnenberg JL, Hada M, Ehara M, Toyota K, Fukuda R, Hasegawa J, Ishida M, Nakajima T, Honda Y, Kitao O, Nakai H, Vreven T, Montgomery JA, Jr, Peralta JE, Ogliaro F, Bearpark M, Heyd JJ, Brothers E., Kudin KN, Staroverov VN, Kobayashi R, Normand J, Raghavachari K, Rendell A, Burant JC, Iyengar SS, Tomasi J, Cossi M, Rega N, Millam JM, Klene M, Knox JE, Cross JB, Bakken V, Adamo C, Jaramillo J, Gomperts R, Stratmann RE, Yazyev O, Austin AJ, Cammi R, Pomelli C, Ochterski JW, Martin RL, Morokuma K, Zakrzewski VG, Voth GA, Salvador P, Dannenberg JJ, Dapprich S, Daniels AD, Farkas O, Foresman JB, Ortiz JV, Cioslowski J, Fox DJ. Gaussian 09. Revision A.02, Gaussian Inc. Wallingford CT. 2009.
- [11] Curtiss LA, Redfern PC, Raghavachari K. Gaussian-4 Theory. *J Chem Phys.* 2007;126:084108-1--12.
- [12] Verevkin SP, Emel'yanenko VN, Notario R, Roux MV, Chickos JS, Liebman JF. Rediscovering the wheel. Thermochemical analysis of energetics of the aromatic diazines. *J Phys Chem Lett.* 2012;3(23):3454-9.
- [13] McQuarrie DA. Statistical mechanics, Harper's chemistry series. New York: Harper & Row; 1976.
- [14] Cox JD, Wagman DD, Medvedev VA. CODATA key values for thermodynamics. New York: Hemisphere Pub Corp; 1989.
- [15] Olofsson G. Assignment of uncertainties. In: Sunner S, Månsson M, editors. Combustion calorimetry. Oxford: Pergamon Press; 1979. p. 137-59.
- [16] Wieser ME, Holden N, Coplen TB, Böhlke JK, Berglund M, Brand WA, De Bièvre P, Groning M, Loss RD, Meija J, Hirata T, Prohaska T, Schoenberg R, O'Connor G, Walczyk T, Yoneda S, Zhu XK. Atomic weights of the elements 2011 (IUPAC technical report). *Pure Appl Chem.* 2013;85(2):1047-78.
- [17] Meier M, Dogan B, Beckhaus H, Ruchardt C. Heats of formation and heats of isomerization of isocyanides. *New J Chem.* 1987;11:1-6.
- [18] Abboud JLM, Jimenez P, Roux MV, Turrion C, Lopez-Mardomingo C. Structural effects on the thermochemical properties of organic compounds. III. Enthalpies of combustion, vapour pressures and enthalpies of sublimation, and standard enthalpies of formation in the gaseous phase, of adamantane-1-carboxylic acid methyl ester and of adamantane-1-carbonitrile. *J Chem Thermodyn.* 1992;24(12):1299-304.
- [19] Abboud JLM, Jimenez P, Roux MV, Turrion C, Lopez-Mardomingo C. Structural effects on the thermochemical properties of carbonyl compounds. I. Enthalpies of combustion, vapour pressures and enthalpies of sublimation, and enthalpies of formation of 2-methylpropanamide, 2,2-dimethylpropanamide, and 1-adamantyl carboxamide. *J Chem Thermodyn.* 1989;21(8):859-65.
- [20] Abboud JLM, Jimenez P, Roux MV, Turrion C, Lopez-Mardomingo C, Podosenin A, Rogers DW, Liebman JF. Interrelations of the energetics of amides and alkenes: enthalpies of formation of N, N-dimethyl derivatives of pivalamide, 1-adamantylcarboxamide and benzamide, and of styrene and its α -, trans- β - and β -, β -methylated derivatives. *J Phys Org Chem.* 1995;8(1):15-25.
- [21] Miroshnichenko EA, Lebedev VP, Matyushin YN. Energy properties of adamantane derivatives. *Dokl Phys Chem.* 2002;382(4):40-2.
- [22] Chickos JS, Acree WE Jr. Enthalpies of sublimation of organic and organometallic compounds 1910-2001. *J Phys Chem Ref Data.* 2002;31(2):537-698.
- [23] Chickos JS, Hosseini S, Hesse DG, Liebman JF. Heat capacity corrections to a standard state: a comparison of new and some literature methods for organic liquids and solids. *Struct Chem.* 1993;4(4):271-7.
- [24] Emel'yanenko VN, Verevkin SP. Benchmark thermodynamic properties of 1,3-propanediol: comprehensive experimental and theoretical study. *J Chem Thermodyn.* 2015;85:111-9.
- [25] Nagrimanov RN, Solomonov BN, Emel'yanenko VN, Verevkin SP. Six-membered ring aliphatic compounds: structure-property relationships in phase transitions. *Thermochim Acta.* 2016;638:80-8.
- [26] Gilbert AS. Entropy-enthalpy compensation in the fusion of organic molecules: implications for Walden's rule and molecular freedom in the liquid state. *Thermochim Acta.* 1999;339:131-42.
- [27] Gobble C, Rath N, Chickos J. The vaporization enthalpies and vapor pressures of some primary amines of pharmaceutical importance by correlation gas chromatography. *J Chem Eng Data.* 2013;58(9):2600-9.
- [28] Foulon M, Amoureux JP, Sauvajol JL, Cavrot JP, Muller M. Different aspects of an interesting glassy crystal: 1-cyanoadamantane. *J Phys C Solid State Phys.* 2000;17(24):4213-29.

- [29] <https://scifinder.cas.org/> Scifinder, Copyright © 2016 American Chemical Society.
- [30] Acree WE Jr, Chickos JS. Phase transition enthalpy measurements of organic and organometallic compounds: sublimation, vaporization and fusion enthalpies from 1880 to 2010. *J Phys Chem Ref Data*. 2010;39(4):043101/1--942.
- [31] Gobble C, Chickos JS, Verevkin SP. Vapor pressures and vaporization enthalpies of a series of dialkyl phthalates by correlation gas chromatography. *J Chem Eng Data*. 2014;59(4):1353-65.
- [32] Suntsova MA, Dorofeeva OV. Use of G4 theory for the assessment of inaccuracies in experimental enthalpies of formation of aliphatic nitro compounds and nitramines. *J Chem Eng Data*. 2014;59(9):2813-26.
- [33] Verevkin SP, Emel'yanenko VN, Pimerzin AA, Vishnevskaya EE. Thermodynamic analysis of strain in the five-membered oxygen and nitrogen heterocyclic compounds. *J Phys Chem A*. 2011;115(10):1992-2004.
- [34] Verevkin SP, Emel'yanenko VN, Diky V, Dorofeeva OV. Enthalpies of formation of nitromethane and nitrobenzene: new experiments vs. quantum chemical calculations. *J Chem Thermodyn*. 2014;73:163-70.
- [35] Verevkin SP, Zaitsau DH, Emel'yanenko VN, Stepurko EN, Zherikova KV. Benzoic acid derivatives: evaluation of thermochemical properties with complementary experimental and computational methods. *Thermochim Acta*. 2015;622:18-30.
- [36] Pedley JB. Thermochemical data of organic compounds. Texas: Thermodynamics Research Center (TRC), College Station; 1994.
- [37] Beckhaus H-D, Dogan B, Verevkin SP, Hädrich J, Rüdhardt C. Dependence of anomeric stabilisation on structure in acetals. *Angew Chem Int Ed*. 1990;29(3):320-1.
- [38] Verevkin SP, Dogan B, Beckhaus H-D, Rüdhardt C. Synergic destabilization by geminal ester groups. *Angew Chem Int Ed Engl*. 1990;29(6):674-5.
- [39] Rakus K, Verevkin SP, Beckhaus HD, Rüdhardt C. Substituent effects on the strength of C-C bonds. 15. Geminal substituent effects.VII. Thermochemistry and thermal decomposition of alkyl-substituted tricyanomethyl compounds. *Chem Ber*. 1994;127(11):2225-34.